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Title: Actinide Isotopic Analysis by Atomic Beam Laser Absorption Spectroscopy

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The isotopic analysis of nuclear materials in the field has important applications in nuclear forensics for non-proliferation. In the area of safeguards, the rapid detection of isotopic signatures and ratios of uranium and plutonium reveals whether or not nuclear materials are as declared for a given facility. In the case of post-detonation debris, the analysis provides information about the origin, processing history, and composition of the fuel prior to detonation, and also the overall fission efficiency of the device. Mass spectrometry remains the gold standard for high-precision, high-sensitivity isotopic analysis of nuclear materials, but its complexity, sample preparation requirements, and utility requirements (e.g. voltage/power, high vacuum, compressed gases, strong acids) make it unsuitable for deployment in the field.

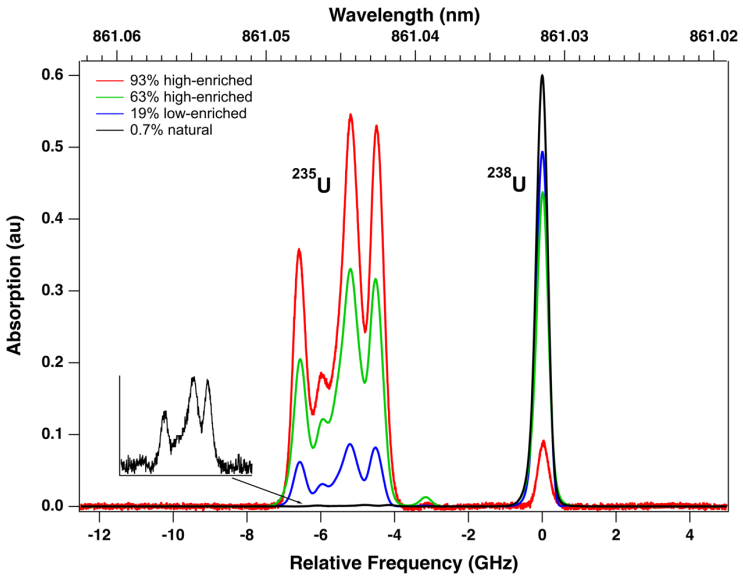

Figure. Isotopically-resolved spectra of uranium at various levels of enrichment.

Chemistry Division researchers have developed new methods and instrumentation for the isotopic analysis of actinides in the field. The method is based on performing laser absorption spectroscopy through a collimated atomic beam of the actinide generated by a high-temperature micro-crucible in rough vacuum. Tantalum micro-crucibles are resistively-heated to temperatures up to 2500 °C. A laser beam from a narrow-linewidth diode laser is tuned across a series of specific isotopic resonances, and the resulting spectrum shows their relative abundance.

Figure. Fieldable prototype instrument for isotopic analysis.

In the case of uranium, for example, the team conducted a series of experiments for the determination of the isotopic composition of a variety of samples of uranium metal, oxides, nitrates, and halides. These samples vary in ^{235}U level of enrichment from natural uranium (0.7%) up to 93%. Excellent signal-to-noise-ratios (up to a few 1000's) are obtained in only a few seconds. By taking the ratio of the areas under each isotope absorption band, the ^{235}U level of enrichment is obtained. The level of enrichment was obtained with an accuracy of 0.2 - 0.4% and a precision of 0.5 - 0.7%. When analyzing oxides and other compounds, the atomic beam is chemically reduced on the fly by addition of lutetium metal to the crucible prior to heating.

References:
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"Isotope-resolved atomic beam laser spectroscopy of natural uranium," Journal of Analytical Atomic Spectrometry **33**, 1862 (2018); doi: 10.1039/c8ja00242h. Authors: Vyacheslav Lebedev, Joshua Bartlett, and Alonso Castro (Actinide Analytical Chemistry, C-AAC).

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